REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

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1. REPORT DATE *(DD-MM-YYYY)* 30-03-2012	2. REPORT TYPE Final Technical Report		3. DATES COVERED *(From - To)* 01-06-2011 to 02-29-2012
4. TITLE AND SUBTITLE MWIR Lasers Using Type II Quantum Well Active Regions on InP Substrates			**5a. CONTRACT NUMBER** FA9550-11-C-0040
			5b. GRANT NUMBER
			5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S) Dr. Igor Kudryashov, Principal Investigator Princeton Lightwave Inc.			**5d. PROJECT NUMBER**
			5e. TASK NUMBER
			5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Princeton Lightwave Inc. 2555 US Route 130 South Cranbury, NJ 08512-5421			**8. PERFORMING ORGANIZATION REPORT NUMBER**
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR 875 N Randolph St Arlington, VA 22203			**10. SPONSOR/MONITOR'S ACRONYM(S)** AFOSR/PKR1
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-OSR-VA-TR-2012-1193

12. DISTRIBUTION / AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT
For this STTR program, we developed modeling for a Type II Quantum Well(QW)-based 2.4-4 μm laser grown on InP substrates that use a unique quantum well design and demonstrated concept feasibility through experimental photoluminescence measurements. The QW design is based on using InGaAs and GaAsSb heterostructures with Type II electronic transitions. This technology enables the potential to manufacture lasers in a broad spectrum, from 2.4-4 μm using Princeton Lightwave Inc's established laser production platform. Our novel quantum well design enhances electron and hole wave-function overlapping, reaching the highest possible material gain. During Phase I of the program, we developed detailed modeling of Type II QW electronic transitions describing optical and gain characteristics using InGaAs and GaAsSb composition layers and demonstrated the concept feasibility through photoluminescence measurements.

15. SUBJECT TERMS
STTR Report

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**	**19a. NAME OF RESPONSIBLE PERSON**
a. REPORT UNCLASSIFIED	**b. ABSTRACT**	**c. THIS PAGE**	UL		**19b. TELEPHONE NUMBER** *(include area code)*

Standard Form 298 (Rev. 8-98)
Prescribed by ANSI Std. Z39.18

Contract No.: FA9550-11-C-0040
Program Title: "MWIR lasers using Type II superlattice active regions on InP substrates"
Contractor: Princeton Lightwave Inc.
Principal Investigator: Igor Kudryashov

Final Report: 30 March 2012

1. Phase I Project Objectives

Our objectives for Phase I of this STTR program were to:
  (1) demonstrate a model that can predict laser optical transition characteristics for wavelengths between 2.4-4µm
  (2) demonstrate by using the model the advantages of using "M" type QW designs compared to "W" type
  (3) Experimentally characterize the optical properties of the grown "M" type QW structure and refine the modeling.

To satisfy Objective (1), we developed theoretical model describing M-type and W-type structures. Model was based on Kane approach taking in account holes and electron bands (8x8 Hamiltonian). Our strategy was: (i) to find optimal realistic parameters for investigated M-type structure based on $Ga_xIn_{1-x}As/GaAs_ySb_{1-y}$ Type-II heterostructures, surrounding by InGaAsP layers, widely used in laser designs on InP substrates, (ii) to calculate gain characteristics for optimal M-structure in wide range of optical transition energies.

Because theoretical model of type II transitions relies on material parameters and results strongly depends on it, we grew an experimental M-type of structure to verify and to tweak our model parameters for higher confidence in theoretical modeling of investigated designs in wide range of material compositions and layer thicknesses. This was one of the goals of Objective 2.

Combining the results obtained for Objectives (1) and (3), we have met Objective (2) by modeling of gain characteristics of M-type structure and W-type structure utilizing the same materials and layer thicknesses as investigated M-type QWs. It allows comparing directly these two types of structures and making conclusion about advantages of M-type structures.

The remainder of this Final Report includes the following sections:
  2.1 Summary Phase I Results
  2.2 Theoretical model
  2.3 Analyze of M-structure parameters
  2.4 Experimental results
In these sections, we address the work performed and results obtained from our program activities.

2. Phase I Results

2.1 Summary of Phase I results

The bulleted list below summarizes the key Phase I achievements:
- Developed theoretical model of optical transitions and gain for M-type and W-type structures.

- Defined on the base of developed model optimal parameters of M-type structure

- Verified theoretical model with experimental data

A detailed discussion of these results is presented in the following sub-sections.

2.2 Theoretical model

We use the effective-mass approximation to model the electron and hole energy levels at Γ-valley. On first stages, we used the six-band Hamiltonian (LK model) for strained semiconductor includes the energy levels from heavy hole (hh), light hole (lh), and spin-orbit split-off (so) bands. The numerical model takes into account the valence band mixing, strain effect. The electron energy band are assumed to be parabolic. The hole energy band is computed via 6×6 diagonalized k·p Hamiltonian matrix, as follows [1]:

	$$H^{\Gamma}_{6\times6}(k) = \begin{pmatrix} H^{U}_{3\times3} & 0 \\ 0 & H^{L}_{3\times3} \end{pmatrix}$$	*(1)*

Here H^{U}_{3x3} and H^{L}_{3x3} are represented as bolow:

	$$H^{\sigma}_{3\times3} = -\begin{pmatrix} P+Q-V_h(z) & R_k \pm iS_k & \sqrt{2}R_k \pm \frac{i}{\sqrt{2}}S_k \\ R_k \mp iS_k & P-Q-V_h(z) & \sqrt{2}Q \pm i\sqrt{\frac{3}{2}}S_k \\ \sqrt{2}Q \mp i\sqrt{\frac{3}{2}}S_k & \sqrt{2}R_k \mp \frac{i}{\sqrt{2}}S_k & P+\Delta-V_h(z) \end{pmatrix}$$ *And* $\sigma = U, L$	*(2)*

where

	$$\begin{aligned} P &= P_k + P_\varepsilon \\ Q &= Q_k + Q_\varepsilon \\ P_k &= \left(\frac{\hbar^2}{2m}\right)\gamma_1(k_t^2 + k_z^2) \\ Q_k &= \left(\frac{\hbar^2}{2m}\right)\gamma_2(k_t^2 - 2k_z^2) \\ S_k &= \left(\frac{\hbar^2}{2m}\right)\sqrt{3}\left(\frac{\gamma_2 + \gamma_3}{2}\right)k_t^2 \\ R_k &= \left(\frac{\hbar^2}{2m}\right)2\sqrt{3}\gamma_3 k_t k_z \\ P_\varepsilon &= -a_v(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \\ Q_\varepsilon &= -\frac{b}{2}(\varepsilon_{xx} + \varepsilon_{yy} - 2\varepsilon_{zz}) \end{aligned}$$	*(3)*

and where $V_h(z)$ is the unstrained valence band edge; $k_t^2 = k_x^2 + k_y^2$, γ_1, γ_2, and γ_3 are the Luttinger parameters; a_v and b are the Bir-Pikus deformation potentials; and $\Delta(z)$ is the spin-orbit split-off energy.

The valance subband structure can be carried out by solving the k·p Hamiltonian matrix, with z axis as the quantization axis. The parameters used such as the band gaps, conduction band offsets, and elastic constants are taken from recent theoretical and experimental results[1,2].

The optical gain of the quantum well was be derived from Fermi's golden rule as below[3]:

$$g(\hbar\omega) = \frac{\pi e^2}{n c \epsilon_0 m^2 \omega L} \sum_{\eta=T,l} \sum_{\sigma=U,L} \sum_{n,m} \int \left| \hat{e} \cdot M_{nm}^{\eta\sigma}(k_t) \right|^2$$
$$\times \frac{\left(f_n^c(k_t) - f_{\sigma m}^V(k_t) \right) \left(\frac{\gamma}{\pi} \right)}{\left(E_{b,\sigma nm}^{cV}(k_t) - \hbar\omega \right)^2 + \gamma^2} \frac{k_t \, dk_t}{2\pi} \qquad (4)$$

where

$$f_n^c(k_t) = \frac{1}{1 + exp\left(\frac{E_n^c(k_t) - F_c}{k_B T} \right)}$$
$$f_{\sigma m}^V(k_t) = \frac{1}{1 + exp\left(\frac{E_{\sigma m}^V(k_t) - F_v}{k_B T} \right)} \qquad (5)$$
$$E_{b,\sigma nm}^{cV}(k_t) = E_n^c(k_t) - E_{\sigma m}^V(k_t)$$
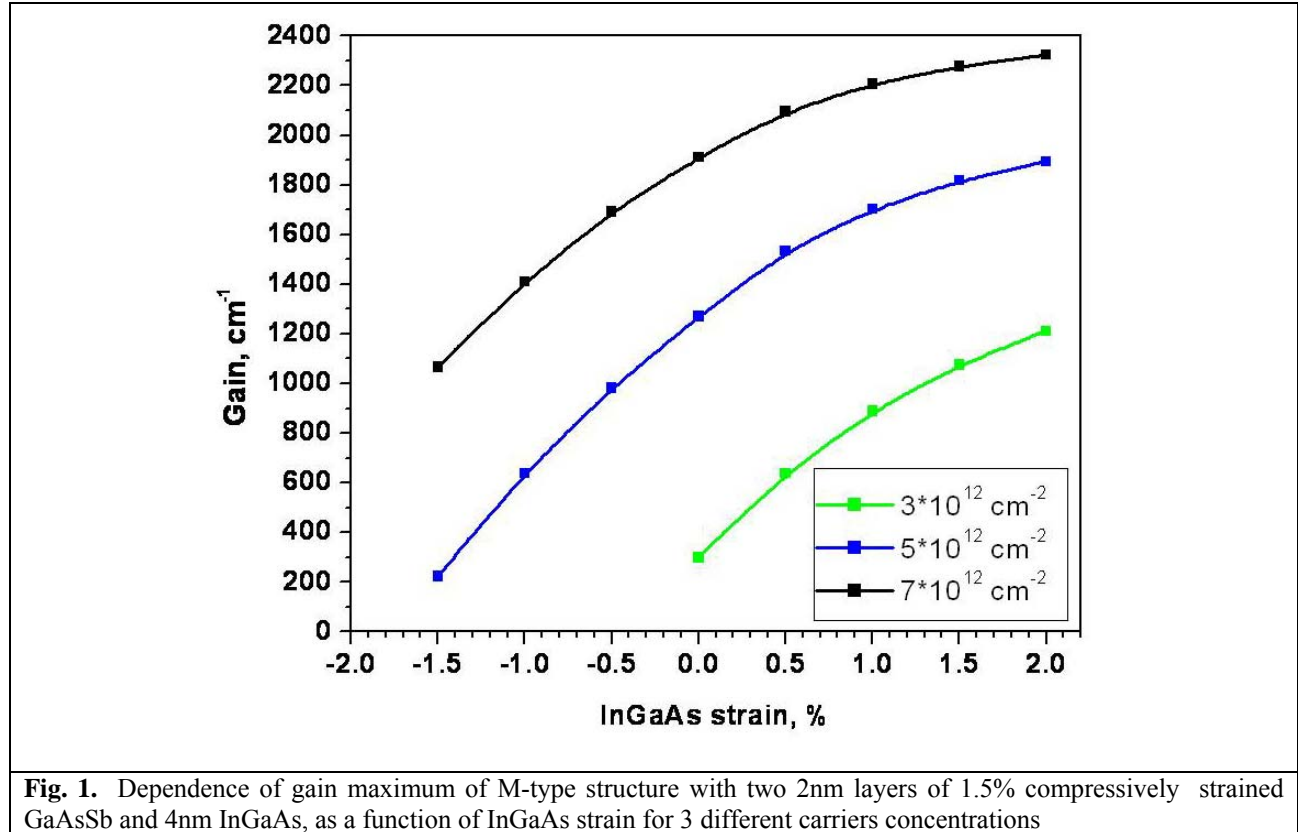
Fc and Fv are the quasi-Fermi levels for electrons and holes, which can be determined by normalizing the total number of electrons or holes. And e is the magnitude of the electron charge, m is the electron mass in free space, $\hat{e}$ is the polarization vector of the optical electric field, n and L are refractive index and well width of the quantum well, γ is the half linewidth of the Lorentzian function, here $\gamma = \hbar/\tau_{int}$, and $M_{nm}^{\eta\sigma}(k_t) = \left\langle \Psi_{m,k_t}^{V\sigma} \left| p \right| \Psi_{n,k_t}^{c\eta} \right\rangle$ is the momentum matrix element where p is the momentum operator. We assume that τ_{int} is a constant value, $\tau_{int} = 1 \times 10^{-13}$ s, and our gain calculation is done on TE mode.

Later we extended our model to Kane model taking in account holes and electron bands (8x8 Hamiltonian).

2.3 Analyze of M-type structure parameters

Main goal is the investigation how parameters of M-structure (thickness of layers, strain and properties of surrounding layers) impact on gain characteristics.

Our initial key results of modeling portray the dependence of gain on strain. The typical dependences of gain maximum on strain in InGaAs layer are shown on Fig.1. The basic M-structure contained two 2nm 1.5% compressively strained GaAsSb layers in between a 4nm InGaAs layer. Three carrier concentrations $3*10^{12}$ cm^{-2}, $5*10^{12}$ cm^{-2} and $7*10^{12}$ cm^{-2} were demonstrated.

Fig. 1. Dependence of gain maximum of M-type structure with two 2nm layers of 1.5% compressively strained GaAsSb and 4nm InGaAs, as a function of InGaAs strain for 3 different carriers concentrations

The main conclusion can be made from these dependences that compressive strain is preferable for M-type structure to achieve high gain.

In framework of developed LK model, we estimated gain and operation wavelength dependence on layer thicknesses as well. Fig.2 shows dependences of gain maximum and operating wavelength for M-structure containing two 2nm of 1.5% compressively strained GaAsSb layers and the thickness of 1.5% compressively strained InGaAs. Increasing the InGaAs layer thickness shifts the operation wavelength toward longer wavelengths. However, maximum gain drops with increasing thickness, because of the reduced overlapping integral. Evidently, the gain value for this structure is still promising even for transitions >3.3µm wavelength. Changing strain of layers and the GaAsSb layer thickness allows extending the wavelength up to 4 µm.
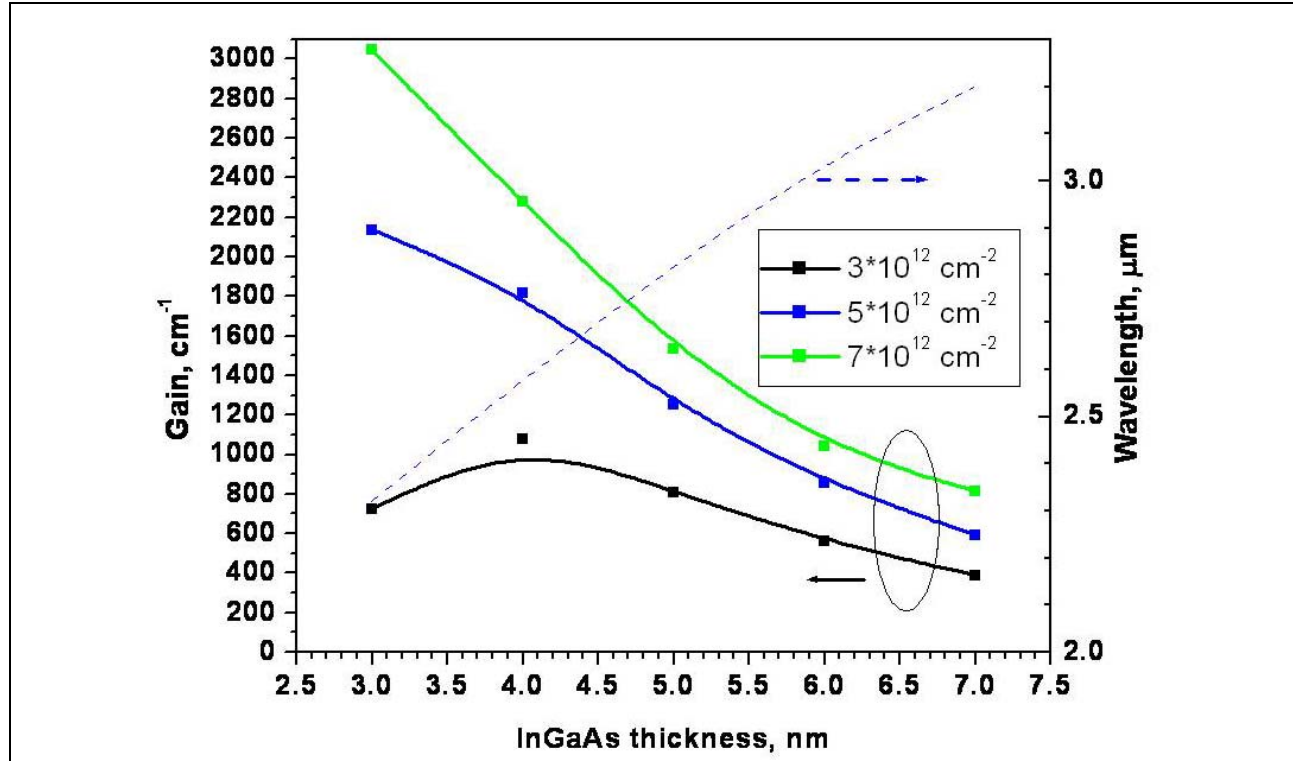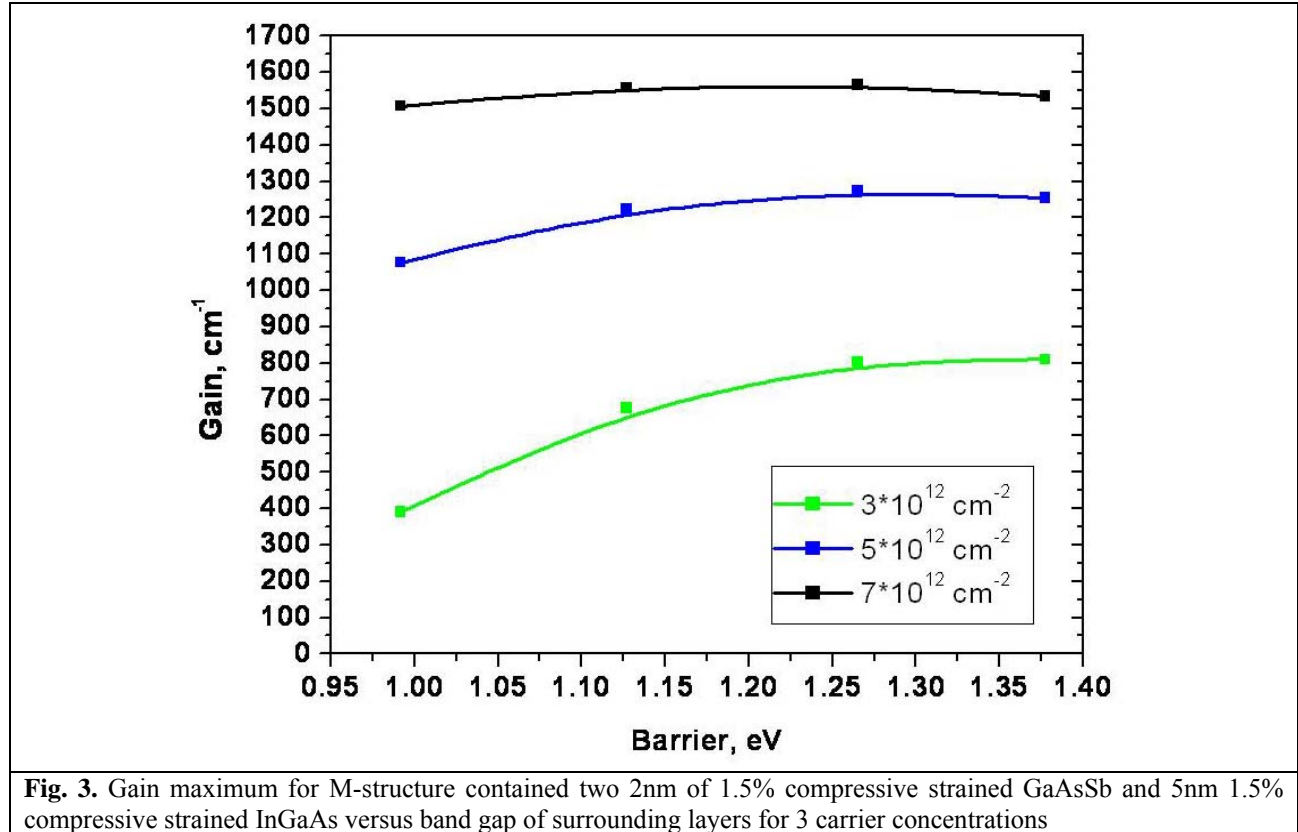
Fig. 2. Dependences of gain maximum and operating wavelength for M-structure with two 2nm layers of 1.5% compressive strained GaAsSb and 1.5% compressively strained InGaAs thickness for 3 different carriers concentrations

   The third goal of the initial modeling was estimation of impacting the surrounding M structure layers on gain characteristics. It is clear from conclusions made before that we should look at tensile strained materials to compensate compressive strain in M-structure. Besides, we should implement surrounding QW structure materials providing significant barriers for holes and electrons to reach high gain characteristics. InGaAsP material is a good candidate for practical realization of the Al-free laser structure based on M-type quantum wells and grown on InP substrate. The impact of quaternary band gap on gain performance is shown in Figure.3. Analysis of these results shows that proper quaternary composition selection is important for high gain performance and the desirable range of band gaps for quaternary has to be between 1.15-1.3eV. We decided to select $In_{0.86}Ga_{0.14}As_{0.15}P_{0.85}$ composition for our first experimental structure and future theoretical investigations. It has a characteristic band gap of ~1.26eV and ~0.5% tensile strain that will allow compensating compressive strain in M-type quantum well.

Fig. 3. Gain maximum for M-structure contained two 2nm of 1.5% compressive strained GaAsSb and 5nm 1.5% compressive strained InGaAs versus band gap of surrounding layers for 3 carrier concentrations

As summary of initial modeling results:
- GaAsSb layers and InGaAs layer in M-type of structure have to be compressively strained to reach the best gain characteristics
- Estimated practical range of layers thicknesses for M-type structure providing optical transitions in 2.4-4um range: 2-3nm of GaAsSb and 4-8nm of InGaAs
- Surrounding of M-type structure layers have to be tensile strained to compensate heavily compressively strained M-type structure.
- Band gap of surrounding layers should be larger than 1.15eV
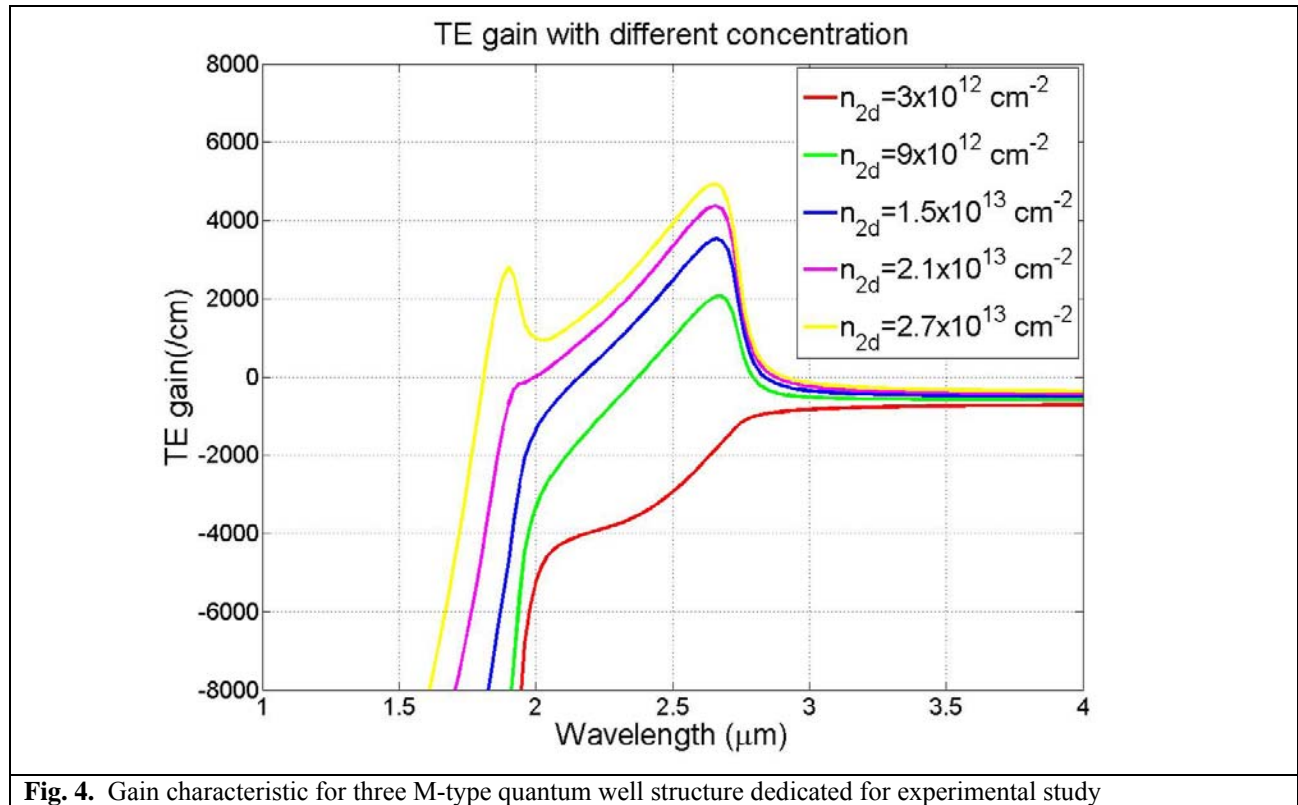
2.4 Experimental results

On the base of conclusions made during initial theoretical investigations of M-type structure, we selected epi-structure satisfied requirements mentioned above. Active region of this structure containing three M-type QWs is shown in Table 1

Table1. Active region of epi-structure

Layer	Material	Thickness ,nm	X	y	Strain, %
Barrier	$InGa_xAs_yP$	20	0.14	0.15	-0.49
QW	$GaAs_{1-y}Sb_y$	2		0.618	1
QW	$In_{1-x}Ga_xAs$	5	0.25		1.5
QW	$GaAs_{1-y}Sb_y$	2		0.618	1
Barrier	$InGa_xAs_yP$	7	0.14	0.15	-0.49

QW	$GaAs_{1-y}Sb_y$	2		0.618	1
QW	$In_{1-x}Ga_xAs$	5	0.25		1.5
QW	$GaAs_{1-y}Sb_y$	2		0.618	1
Barrier	$InGa_xAs_yP$	7	0.14	0.15	-0.49
QW	$GaAs_{1-y}Sb_y$	2		0.618	1
QW	$In_{1-x}Ga_xAs$	5	0.25		1.5
QW	$GaAs_{1-y}Sb_y$	2		0.618	1
Barrier	$InGa_xAs_yP$	20	0.14	0.15	-0.49

We used extended theoretical modeling to estimate gain characteristics for this structure. This model took in account holes and electron bands (8x8 Hamiltonian). Figures 4 shows result of this modeling. Remarkable, that new model predicts significantly different gain spectrum for investigated structure in comparison with previous calculations , but maximum of gain value per one QW is close to previous calculations.

Fig. 4. Gain characteristic for three M-type quantum well structure dedicated for experimental study

Metal-Organic Chemical Vapor Deposition (MOCVD) technology were used to grow experimental heterostructure. Growth temperature was about 525ºC for M-type regions and brief pauses will be introduced between layers to stabilize the group-V flux. As we have determined from previous experiments that such short pauses lead to reasonably abrupt and high quality interfaces.
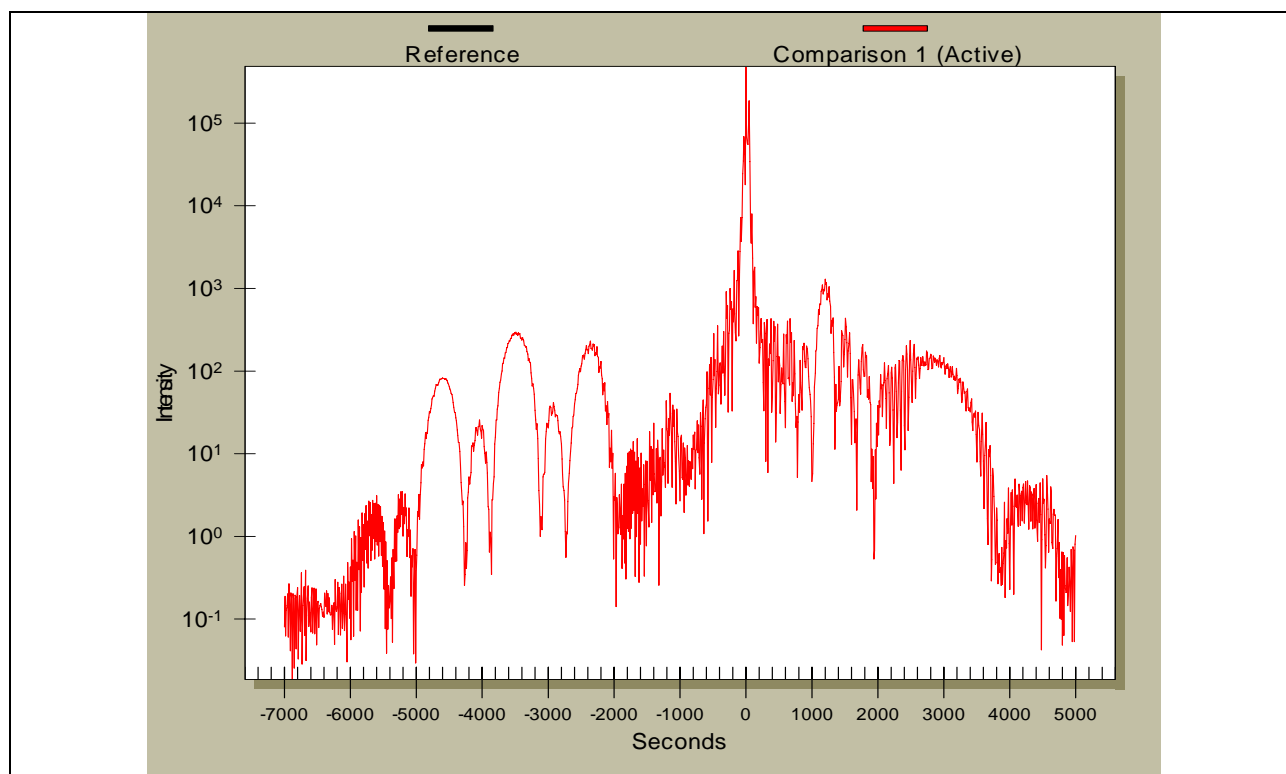
Fig. 5. Simulated spectra of X-ray diffraction of grown epi-structure.
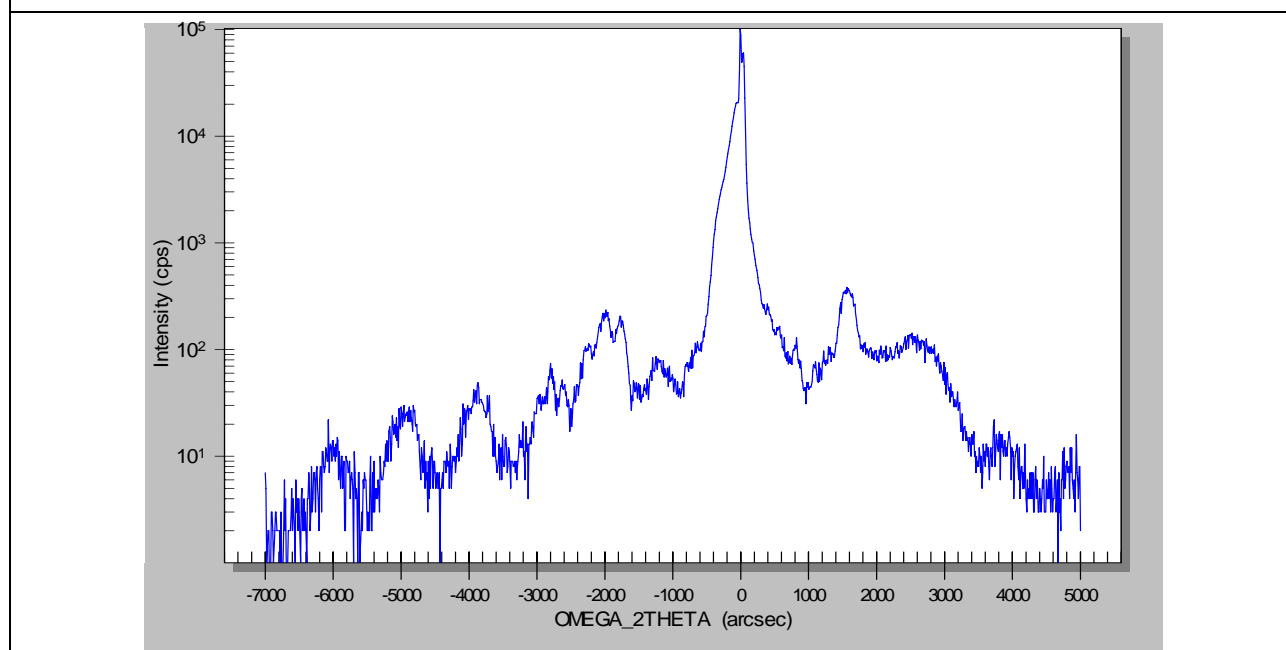

Fig. 5. Experimental spectra of X-ray diffraction of grown epi-structure.

X-rays diffraction measurements demonstrate high quality of grown material. Figure 4 shows simulated diffraction and Figure 5 experimental one. But there is a sign for possible small

interdiffusion between GaAsSb and InGaAs materials in QW region. Evidence of it can be gotten from comparison simulated and experimental dependences on angle in a range from -6000 to -2000 arcsec. The dependence in this range reflects properties of multi quantum wells region in epi-structure. Simulated curve demonstrates series of peaks with different amplitudes in this region. Experimental dependence is characterized by number of peaks with similar amplitudes. But effect inter-diffusion is small and total quality of grown material allows us to use spectral properties of this structure to refine our theoretical model.
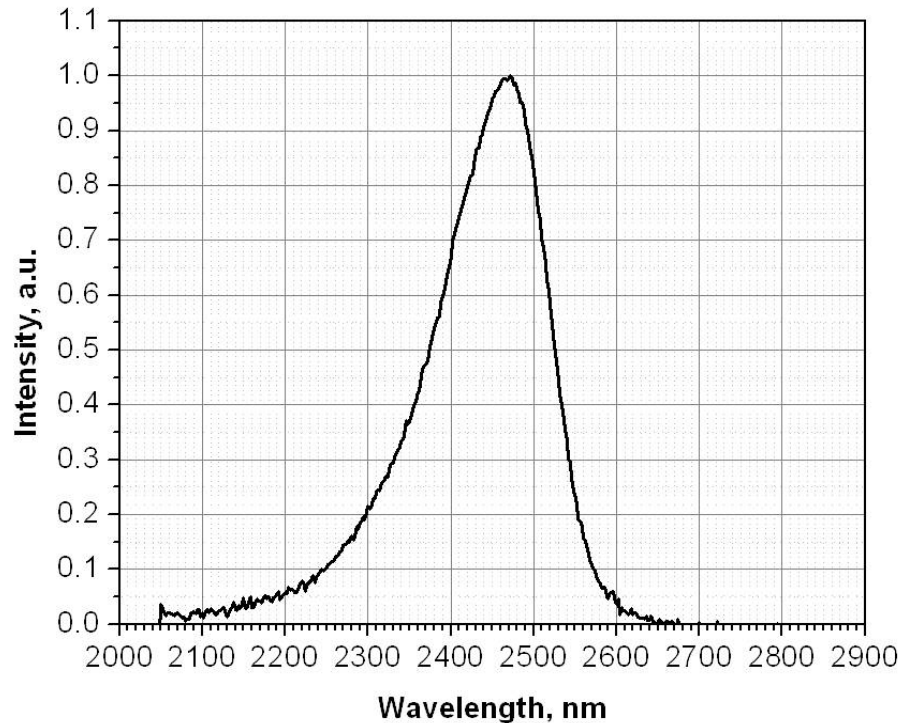

Fig. 6. Room temperature photoluminescence of grown epi-structure.

Strong room temperature photoluminescence (PL) at ~2.5μm was observed at optical excitation by 0.8μm laser diode (Figure 3). Observed spectral position of PL is slightly shorter than it was predicted by our theoretical model.

The main impact on spectral position in developed model has band offset parameter between GaAsSb and InGaAs. We used interpolation formulas taken from [1] in our theoretical model. The value calculated on the base of these formulas is ~0.485eV. We need to decrease this offset on 80meV to get good agreement with experiments. This adjustment looks reasonable for us, because of it is known that interpolation formulas work well for unstrained compositions, where there are a lot of experimental data, but do not work well for high strained composition. Gain characteristics for investigated structure calculated with experimental feedback is shown on Figure 7. These results are very promising for the practical realization of the laser device.
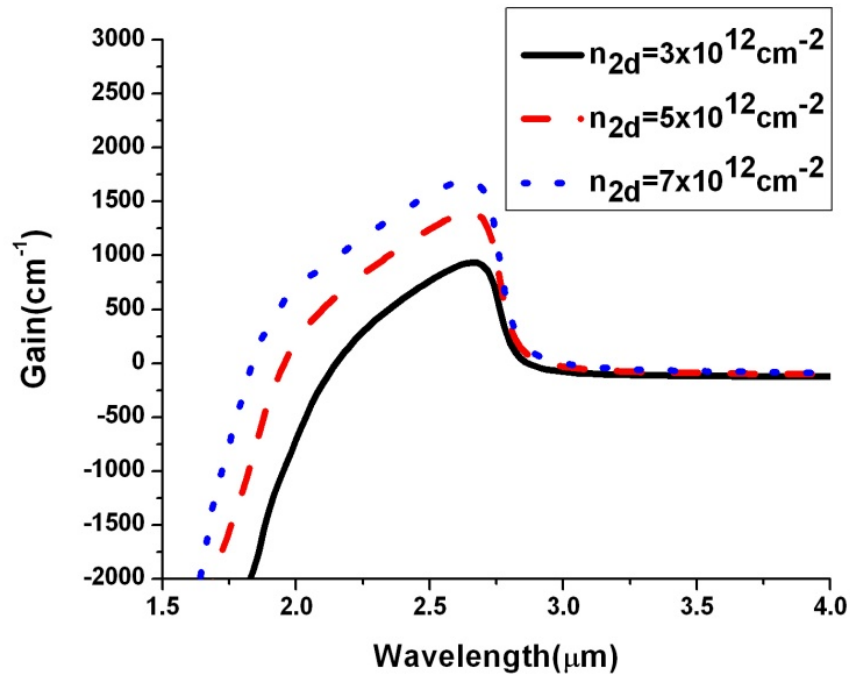
Fig. 7. Gain characteristic for M-type quantum well structure dedicated for experimental study.

3. Researchers involved in Phase I program activities

At Princeton Lightwave:

        Dr. Igor Kudryashov, Principal Investigator

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        Dr. Victor Khalfin, Staff Engineer

At University of Virginia

        Prof. Archie Holmes

        Baile Chen, PhD Candidate

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